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ORIGINAL ARTICLE

Assessment of a degree of geochemical maturation and activity of a closed landfill site in Poland

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Abstract The study was conducted around a reclaimed municipal landfill site, located in the suburbs of Otwock, about 25 km south-west of Warsaw (Poland). The research was carried out about 15 years after the landfill site ceased operation. According to the regulations on landfills sites in Poland (J. of Laws of 2002 No. 220 item 1858, Regulation of the Minister of Environment of 9 December 2002 on the scope, time, manner and conditions of landfill monitoring; J. of Laws of 2010 No. 238 item 1588, Regulation of the Minister of Environment of 8 December 2010 amending the regulation on the scope, time, manner and conditions of landfill monitoring), the monitoring period will end in 2021. Despite reclamation, the landfill site still affects the environment. The aim of this study was to assess the geochemical maturation of the stored waste and the landfill site activity by determining the current phase of the landfill site lifetime. This was achieved by means of non-standard application of carbon stable isotopes for the assessment of the landfill activity combined with conventional methods for measuring COD, BOD, DOC, BOD/COD ratio, and H_2S and O_2 content in biogas. An analysis of carbon isotopic composition and COD, BOD, DOC of the leachate-contaminated groundwater and O_2 concentration in biogas indicated that the landfill site was between the methanogenic phase and air intrusion phase. However, BOD/COD ratio and H_2S concentration in the biogas suggested that the landfill site is still active and the wastes contain readily biodegradable organic matter. The impact of the landfill site on the environment seems quite significant. In this case, and in other situations when the impact of a landfill site on the environment is high after the post-closure stage, the monitoring period should be extended.

Keywords Reclaimed municipal landfill site · Post-closure stage · $\delta^{13}C_{DIC}$ · Hydrogen sulfide · Methanogenic phase · Air intrusion phase

Introduction

Despite waste sorting schemes and partial recycling, a considerable amount of waste is stored in the landfill sites. According to the data of the Central Statistical Office (Environmental Protection 2013), in 2012 there were 527 municipal landfill sites operating in Poland, with a total area of 2197.6 ha, with 12,085,000 t of wastes. At this time, ten of these landfill sites, with total area of 132.1 ha, ceased to accept wastes and entered a post-closure stage. According to the Polish regulations (J. of Laws of 2002 No. 220 item 1858; J. of Laws of 2010 No. 238 item 1588), the post-closure stage is a period of 30 years, counted from the day of the landfill closing decision. During this period, biogas and leachate production must be monitored every 6 months. According to this regulation, the post-closure stage for the Otwock landfill site covers a period from 1991 to 2021. The research presented here was conducted in the years 2006 and 2007, thus reflecting the second decade of the post-closure stage, and more precisely the period of about 15 years after the landfill site closing. However, the landfill site still exerts a considerable impact on the environment. In Otwock, and in other cases where the impact of

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the landfill site on the environment is still high after the post-closure stage, the monitoring period should be extended and the measurements should be continued.

As reported in the literature, the environmental risks posed by landfill sites are present for a long time during the post-closure stage, even after the site reclamation. As a result, a biogas causing the greenhouse effect and a leachate contaminating groundwater in the aquifer are formed. This is a serious problem, a solution to which is sought after in many countries. There are attempts at converting the negative phenomenon of methane formation into a positive one, by harvesting and using the gas for economic purposes. In Poland, energy recovery from the biogas is not very popular, as it is often not economically viable, which can be relatively easily estimated (e.g. Zacharof and Butler 2004). Biogas is used for electricity production in 58 cases, and for heating in only 13 cases (Environmental Protection 2013). In Otwock, passive venting of the landfill site is applied, allowing biogas to escape to the atmosphere. In small landfill sites, where biogas harvesting is usually not cost-effective, this is another solution employed to prevent negative effects of the biogas emission. It facilitates stabilization and aerobic decomposition of the deposited wastes. The aeration leads to an enhanced aerobic degradation of the landfilled waste, also referred to as aerobic in situ stabilization. For this purpose, in situ aeration of the wastes and laboratory tests are practiced (Ritzkowski and Stegmann 2012, 2013; Hrad et al. 2013). In-situ aeration is a conventional technique of reclamation applied to soils contaminated with hydrocarbons ("bioventing"). It has recently been applied in order to reduce residual methane emissions from old and closed landfill sites in some European countries, including Germany or Italy.

Due to a growing concern for the natural environment, including air, soil, surface water and groundwater, closed landfill sites are reclaimed and monitored (J. of Laws of 2002 No. 220 item 1858; J. of Laws of 2010 No. 238 item 1588), (Manczarski and Lewicki 2012), and newly constructed landfill sites must meet stricter criteria (J. of Laws of 2003 No. 61 item 549; Klimek et al. 2010). The environmental impact of the currently constructed landfill sites can be markedly minimized through the use of appropriate methods (Klimek et al. 2010). Serious problems are posed by old landfill sites, created in the 1960s, without appropriate engineering structure (no bottom liner), often located in former mining pits or natural depressions, where non-selected wastes were collected in an uncontrolled manner and documentation was insufficient. Even many years after closing and reclamation, such landfill sites continue to negatively affect the environment, as shown on the example of the Otwock landfill site. Assessing further activity of such facilities and predicting changes within them is almost impossible. Only current biogas composition and groundwater quality can serve as indicators of the landfill site lifetime phase, but they do not allow for predicting any future changes. At the Otwock landfill site, the difficulties in assessing its activity are due to not only the complexity of hydrogeological conditions (part of the landfill base is located within the aquifer), but mainly to a lack of documentation on the waste storage (amount and type of deposited materials), because the landfilling was started as a dump. Moreover, biodegradation of synthetic polymers, recognized as major solid environmental pollutants, is difficult to assess, because it depends on both the environment in which they are placed and the chemical properties of the polymer (Sharma and Mudhoo eds. 2011).

Landfill activity and the maturation of the stored wastes can be assessed using various methods. The following parameters are typically analyzed to determine the landfill phase: pH, BOD, COD, DOC, and heavy metals (Christensen and Kjeldsen 1989, 2001; Kjeldsen et al. 2002; Kulikowska and Klimiuk 2008). The usefulness of carbon isotope determination in demonstrating environmental impact of a landfill site was reported by North et al. (2006). There are many classification phases for landfill sites (e.g. Kjeldsen et al. 2002), but in this paper a division into six phases was adopted: (1) aeration and acid phase, (2) initial methanogenic phase, (3) stable methanogenic phase, (4) air intrusion, (5) CO_2 phase/aerobic, (6) background, as proposed by Wimmer et al. (2013). This classification is based on the changes of carbon isotopic signature during each of the landfill lifetime phases. According to this classification, during the aeration and acid phase at the beginning of the landfilling, $\delta^{13}C_{DIC}$ in the landfill leachate is between −20 and −25 ‰. With the beginning of methane production in the initial methanogenic phase, $\delta^{13}C_{DIC}$ is increased and reaches a maximum of about +15 ‰ during stable methanogenic phase. Natural intrusion of oxygen, starting at the end of the methanogenic phase, moves the isotopic signature again towards negative values. In the carbon dioxide phase, $\delta^{13}C_{DIC}$ ranges from −20 to −25 ‰, as the result of aerobic degradation of organic matter in the waste.

The aim of this study was to assess the landfill activity by determining the phase in which the landfill site currently is. To meet the study objective, a non-conventional method was used, involving an analysis of isotopic composition of inorganic carbon in the water from a piezometer located in the closest vicinity to the landfill site and exhibiting the greatest contamination with the leachate, as well as an analysis of oxygen and hydrogen sulfide in the biogas. Measurements of BOD, COD and DOC were used as parameters supporting the result interpretation. The chemical composition of leachate was not analyzed, because at the time of sampling, the leachate collector was empty and it was impossible to collect any samples.

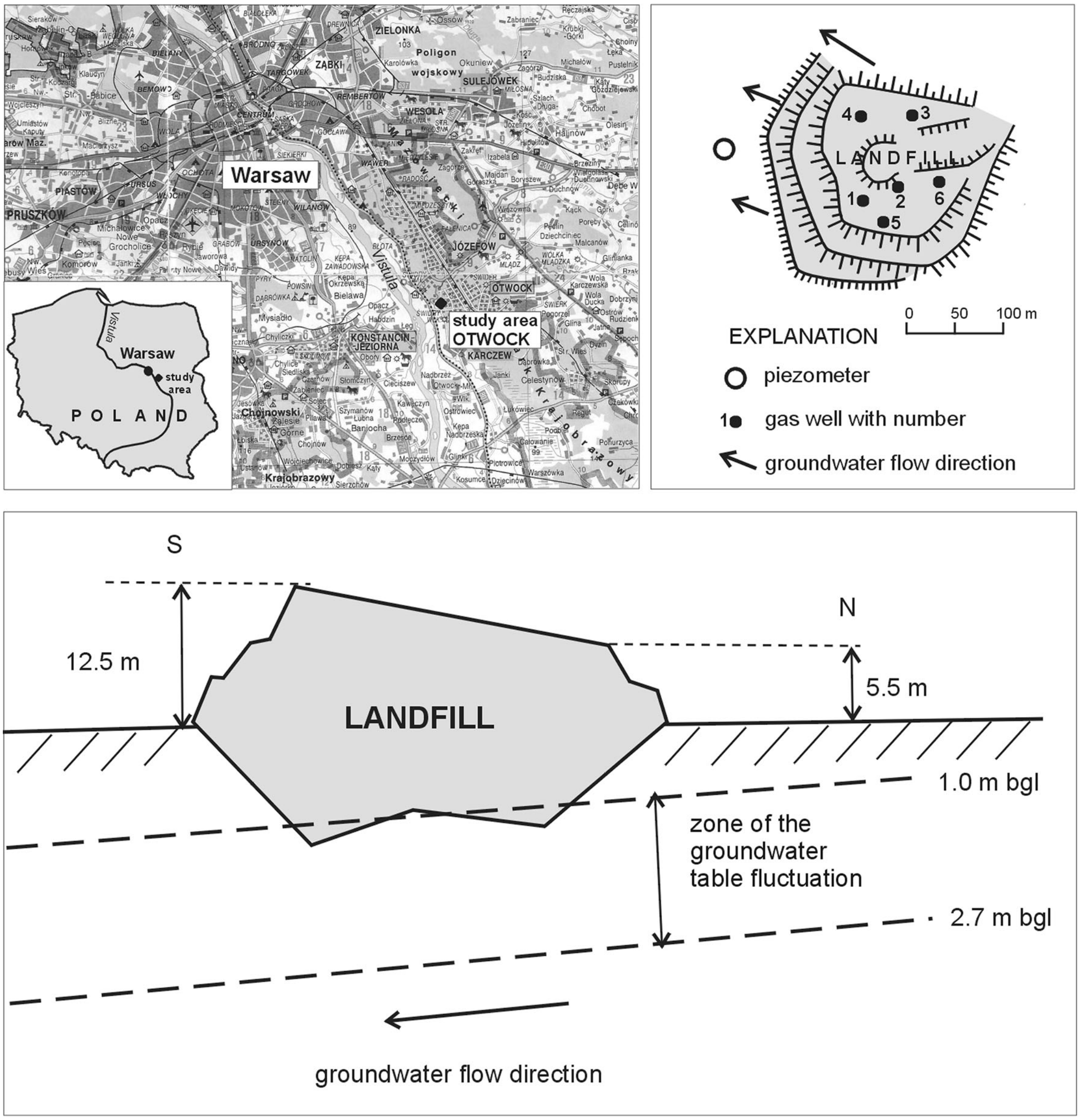

Fig. 1 Location of the study area, arrangement of the sampling points, and the site hydrogeological conditions

Research site description

The study was conducted around the reclaimed municipal landfill site, located in the suburbs of Otwock, about 25 km south-west of Warsaw (Poland) (Fig. 1). The landfill site began to receive wastes in 1961 and continued to do so for a period of almost 30 years. It was closed in 1991. According to the literature (e.g. Kjeldsen et al. 2002; North et al. 2006), the Otwock landfill site can be classified as an old landfill, for which the period of waste storage exceeded 10 years. Many years have passed since then but biogas and leachate are still being generated and waste decomposition is in progress. The Otwock landfill site covers an area of 2.8 ha and it received municipal waste from the nearest community: Otwock, Karczew, Józefów, Wiązowna and Celestynów. The amount and type of waste in the Otwock landfill site is not known (it is not well documented), because landfilling was uncontrolled. After closing, the landfill site was covered with a layer of soil, consisting of approximately 30–50 cm of compost, and planted with grass. North slope of the landfill site is 12.5 m high, while the south slope is lower, only about 5.5 m (Fig. 1). The landfill site is surrounded by a leachate collection trench. Eight evenly spaced gas wells were installed for passive venting of the waste. Currently, there are only six gas wells, in which the measurements were performed, as two of them had been destroyed.

The landfill site is located on the floodplain terrace (Holocene), represented by the river Vistula's highly permeable deposits—gravels and varigrained sands, locally containing organic matter distributed within the aquifer. Hydraulic conductivity of the aquifer ranged from 6.5×10^{-4} to 1.6×10^{-3} m/s, depending on the clay content. Groundwater flow velocity, determined by Małecki eds. (Malecki 2006), is expected to be about 70 m per year. During the periods of high recharge and high river levels (flood peaks in the early spring), most of the landfill body resides below groundwater table, which is in general less than 2.7 m below the ground surface. In the research period, groundwater table ranged from 1.5 to 2.7 m bgl (from May to December) and from 1.0 to 1.7 m bgl (in March), depending on the sampling site, but the general groundwater flow direction did not change.

Analytical procedures

The study analyzed carbon isotope composition and concentrations of dissolved organic carbon, biochemical and chemical oxygen demand in the groundwater as well as the concentration of hydrogen sulfide and oxygen in the biogas. Analysis of groundwater was performed in a piezometer located closest to the landfill site and showing the greatest contamination by leachate. The measurements of biogas were performed in six gas venting wells installed at the landfill site (Fig. 1). Chemical composition of the leachate was not analyzed, as the leachate collector was empty and it was impossible to collect any samples.

Laboratory research of the groundwater and field measurements of biogas were conducted simultaneously every quarter from May 2006 to March 2007 (25 May 2006, 23 August 2006, 8 December 2006, and 6 March 2007), using the same research equipment and analytical procedures (Weight and Sonderegger 2000; Witczak et al. 2013). Gas content in the landfill site was evaluated six times, with additional measurements on 11 July 2006 and 6 August 2013. Temperature and pH of the groundwater were measured in the field using a pH-meter 330i (WTW). Samples for carbon isotopic composition $\delta^{13}C_{DIC}$ were collected in 1000 ml glass bottles (Simax Corp.). Water samples were fixed with $SrCl_2$–NH_4OH. Water collected and analyzed for carbon isotopes was treated following the methods described in a previous article (Porowska and Leśniak 2008). Carbon isotopic composition was determined using a Finnigan MAT Delta+ mass spectrometer at the Isotope Laboratory of the Institute of Geological Sciences, Polish Academy of Sciences in Warsaw. $\delta^{13}C$ values of the dissolved inorganic carbon samples were reproducible to ±0.1 ‰. They were expressed relative to the VPDB standard. Overall precision of routine $\delta^{13}C_{DIC}$ measurements exceeds 0.1 ‰.

Concentration of dissolved organic carbon (DOC), biochemical oxygen demand (BOD), and chemical oxygen demand (COD) in the groundwater samples was assessed at the Central Chemical Laboratory of Polish Geological Institute—National Research Institute in Warsaw. Hydrogen sulfide and oxygen content in the biogas were measured in the field using gas tube detectors "Gastec" manufactured by Gastec Corporation of Japan (Gastec Handbook 2004).

Results

Numerous biogas and groundwater components were analyzed for the Otwock landfill site but the paper presents only the parameters directly related to the assessment of the landfill site geochemical maturation and its activity. pH of the groundwater measured in the field ranged from 7.32 to 7.76 (Table 1). $\delta^{13}C_{DIC}$ varied from +3.6 to −3.5 ‰ (Fig. 2). Single measurements of BOD and COD were performed in July, and their values reached 82 and 100 mg O_2/dm^3, respectively. BOD/COD ratio was 0.82. Low DOC concentrations, from 5.8 to 13.5 mg/dm^3, were found in the research period.

Concentration of hydrogen sulfide measured in the gas wells revealed considerable temporal and spatial variability. It varied from non-detectable (<0.25 ppm) in July to 47 ppm in December and March (Table 2). In the samples collected in May 2006, the highest content of hydrogen sulfide was only 8 ppm, while in July 2006 H_2S content

Table 1 Field and laboratory measurements of the selected components of leachate-contaminated groundwater

Component	Unit	Date of measurement			
		25.05.2006	11.07.2006	8.12.2006	6.03.2007
pH	(–)	7.56	7.32	7.67	7.73
$\delta^{13}C_{DIC}$	(‰)	+2.9	+2.0	+3.6	−3.5
BOD	(mgO_2/dm^3)	n.a	82	n.a	n.a
COD	(mgO_2/dm^3)	n.a	100	n.a	n.a
DOC	(mg/dm^3)	11.0	13.5	5.8	6.5
BOD/COD	(–)	n.a	0.82	n.a	n.a

n.a not analyzed

was below the detection limit (<0.25 ppm), except for the well No. 4, where it was 4 ppm. Subsequent sampling periods indicated a marked increase in H_2S concentration.

The highest H_2S concentration in August was 23 ppm, while in December and March it reached up to 47 ppm. Verification studies carried out in August 2013 showed that hydrogen sulfide was still formed in the landfill site, and its content ranged from 5 to 12 ppm.

In May 2006, oxygen concentration in the biogas ranged from 1 to 12 % (Table 2). The results from July 2006 showed a decrease in oxygen content to less than 0.5 % in the gas wells 1, 4 and 5, and a clear increase up to 19 % in the gas well No. 6. On the next three sampling days (23 August 2006, 8 December 2006, 3 March 2007) and the follow-up sampling day of 6 August 2013, oxygen content within the entire landfill site was below the detection limit of 0.5 %.

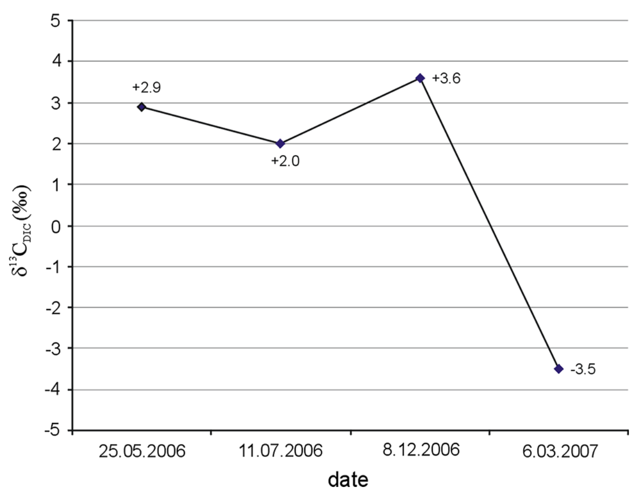

Fig. 2 Carbon isotope composition of groundwater in piezometer

Discussion

Landfill activity and the geochemical maturation of the stored wastes can be assessed using various methods. Usually, the assessment of the landfill phase is based on the following parameters: pH, BOD, COD, DOC, and heavy metals, the concentration of which depends on the landfill age and may change over time. Another method is an assessment based on the biogas composition and concentration of its elements (Christensen and Kjeldsen 1989; Kjeldsen et al. 2002). Although such evaluation seems easy in theory, it is in fact quite complex. This is due to numerous and diverse processes occurring in the landfill site and their variability depending on a number of difficult to assess natural factors, such as moisture and temperature. It is further complicated by the presence of anthropogenic factors. The most problematic issue, especially in old landfill sites that had been started as illegal ones (dumps), is the amount and composition of individual types of waste and a degree of their decomposition in different areas of the landfill site.

In Otwock, the landfill activity analysis included leachate contaminated groundwater, and not the leachate, in which the concentrations of individual contaminants are clearly higher. The landfill stage, indicating its activity, was assessed by determining the inorganic carbon isotope $\delta^{13}C_{DIC}$ in the groundwater from the piezometer located in the closest vicinity to the landfill site and showing the highest share of the leachate. The assessment of the Otwock landfill activity was supplemented with the analysis of BOD, COD and DOC in the water from this piezometer and simultaneous determination of oxygen and hydrogen sulfide content in the biogas harvested from the landfill site.

Table 2 Field measurements of the selected gases in the biogas

Gas well no.	Content of gas	Date of measurement					
		25.05.2006	11.07.2006	23.08.2006	8.12.2006	6.03.2007	6.08.2013
1	H_2S (ppm)	2	<0.25	23	4.5	5	5
2		4	<0.25	16	12	6	8
3		8	<0.25	20	8	23	12
4		8	4	10	8	11	8
5		2	<0.25	16	13	11	6
6		3	<0.25	7	47	47	10
1	O_2 (%)	1	<0.5	<0.5	<0.5	<0.5	<0.5
2		1	4.2	<0.5	<0.5	<0.5	<0.5
3		4.5	16	<0.5	<0.5	<0.5	<0.5
4		6	<0.5	<0.5	<0.5	<0.5	<0.5
5		10.5	<0.5	<0.5	<0.5	<0.5	<0.5
6		12	19	<0.5	<0.5	<0.5	<0.5

Analysis of carbon isotopic composition in the water collected from the piezometer located the closest to the landfill site showed that $\delta^{13}C_{DIC}$ concentration depended on the season and ranged from +3.6 ‰ in the fall to −3.5 ‰ in the winter (Table 1). A comparison of these data with changes in $\delta^{13}C_{DIC}$ concentration over time, proposed by Wimmer et al. (2013), indicates that the landfill site is on the verge of the stable methanogenic phase and air intrusion phase. In the stable methanogenic phase, $\delta^{13}C_{DIC}$ adopts positive values. Air injection during the intrusion phase causes a shift in the isotope signature towards lower values (even negative ones). However, it should be remembered that the analyses were performed for the groundwater heavily polluted with the leachate, and not for the leachate itself, and thus the isotopic composition in the samples differed from a typical leachate. It is estimated that for $\delta^{13}C_{DIC}$ of +3.6 ‰ the landfill impact on the groundwater amounts to 80 %, and it is reduced to 62 % for $\delta^{13}C_{DIC}$ of −3.5 ‰ (Porowska 2015). The estimated $\delta^{13}C_{DIC}$ in the leachate was +11.5 ‰ indicating a stable methanogenic phase. This means that even after many years the activity of the landfill site is still significant, resulting in the formation of leachate contaminating the surrounding groundwater. Previous investigations showed that the values of $\delta^{13}C_{DIC}$ in uncontaminated groundwater around the Otwock landfill site varied between −20.6 and −12.4 ‰ (Porowska 2015). Therefore, the difference between the isotopic composition of dissolved inorganic carbon in natural and leachate-contaminated water is significant.

The results presented below indicate that the landfill site is on the verge of the stable methanogenic phase and air intrusion phase, and not in the initial methanogenesis phase, when the carbon isotopic signature could be similar. The first argument in favor of this claim is that the methanogenesis phase is characteristic for the initial period of waste disposal, lasting from three months to three years (Christensen et al. 1996). Therefore, the discussed landfill site, where the waste disposal began over 50 years ago (the site operated in the years 1961–1991) cannot be in the initial methanogenesis phase. The second argument refers to the concentrations of BOD and COD in the water from the investigated piezometer, amounting to 82 and 100 mg O_2/dm^3, respectively, i.e. much lower than typical ranges of these parameters proposed by Kjeldsen et al. (2002) for the leachate in the initial methanogenesis phase, where mean BOD was 13,000 mg O_2/dm^3, and mean COD was 22,000 mg O_2/dm^3. BOD and COD in the contaminated groundwater in Otwock were similar, but still lower than typical values for stable methanogenic phase, for which mean BOD is around 180 mg O_2/dm^3, and mean COD about 3000 mg O_2/dm^3 (Kjeldsen et al. 2002). It should be pointed out that if the leachate analyses were possible in Otwock, the values of both parameters would be respectively higher, corresponding rather to stable methanogenic phase.

Another argument supporting the hypothesis that the landfill site is on the verge of the stable methanogenic phase and air intrusion phase is temporally and spatially changing oxygen concentration in the biogas. In the initial period of the study, oxygen concentration was as high as 19 %, but in the fall and winter, it dropped below the detection limit of 0.5 % (Table 2). This situation leads to two important conclusions. Firstly, the presence of atmospheric oxygen indicates the beginning of the air intrusion phase, and secondly, the lack of oxygen suggests periodically active methanogenesis processes, typical for the stable methanogenic phase.

The presented analysis shows that the landfill site in Otwock is on the verge of the stable methanogenic phase and air intrusion phase. Depending on local conditions (temperature and moisture), hydrochemical processes occur with varying intensity, causing the formation of biogas and leachate of varying composition. Other literature data on this site suggest that the intensity and direction of changes clearly correlate with the water balance of the landfill body, as described in details in previous publications (Porowska and Gruszczyński 2006, 2013; Porowska 2014).

Another parameter used for the assessment of a landfill age and activity is hydrogen sulfide. It is much more abundant in new landfill sites than in those operating for many years (Kim 2006). Given the period of the Otwock landfill operation, it can be classified as an old facility, and therefore low share of hydrogen sulfide in the biogas is expected.

A comparison of this gas concentration in old and new facilities showed that H_2S content in the biogas measured in Otwock was medium but closer to the new and active landfill sites (Fig. 3). Such a trend was established for the years 2006–2007 and it was confirmed by the results from 2013 (Table 2). Hydrogen sulfide content in the biogas corroborated the conclusions on landfill activity drawn on the basis of groundwater analyses.

Despite the expected end of the stable methanogenic phase and restitution of natural conditions, hydrogen sulfide content in the biogas amounted to 5–12 ppm, thus indicating considerable landfill activity even 24 years after closing.

Landfill activity is a result of organic substance biodegradation and can be evaluated on the basis of BOD/COD ratio (Renou et al. 2008; Kjeldsen et al. 2002). In the leachate from old landfill sites, BOD/COD ratio does not exceed 0.1. This is due to the fact that the biodegradable organic chemicals include humic and fulvic-like compounds, characterized by slower degradation rate than the

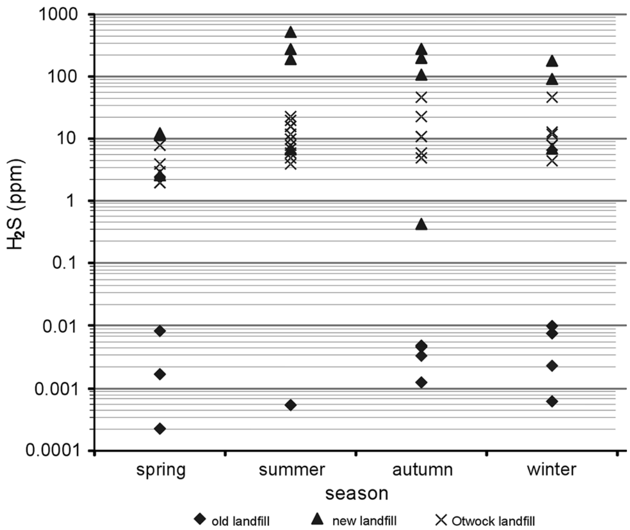

Fig. 3 Hydrogen sulfide concentration in the biogas harvested in Otwock and in new and old landfills (H_2S in old and new biogas after Kim 2006)

organic compounds (mainly volatile fatty acids), undergo quick biodegradation at the initial stage of waste disposal. In Otwock, BOD/COD ratio is much higher than 0.1 (up to 0.82), suggesting the presence of organic compounds that may undergo relatively easy and rapid biodegradation. When the organic matter in the leachate is readily biodegradable, the BOD/COD ratio exceeds 0.4 (Christensen et al. 2001).

DOC values in the contaminated groundwater, ranging from 5.8 to 13.5 mg/dm^3, indicate natural-like conditions. However, it should be pointed out that this parameter is respectively higher in the leachate. DOC concentration in the leachate from old landfill sites ranges from 100 to 1000 mg/dm^3 (McBean et al. 1995).

Conclusions

The presented analysis shows that the landfill site in Otwock, about 15 years after closing, continues to affect its environment. Non-conventional application of carbon stable isotopes $\delta^{13}C_{DIC}$, combined with conventional methods based on COD, BOD, DOC, BOD/COD ratio, as well as H_2S and O_2 concentration in the biogas seemed very useful in the assessment of the degree of geochemical maturation and the landfill activity. Given the carbon isotopic composition $\delta^{13}C_{DIC}$ and COD, BOD, DOC values in the leachate-contaminated groundwater and O_2 in the biogas, it can be concluded that the landfill site is between the methanogenic phase and air intrusion phase. However, BOD/COD ratio, as well as H_2S concentration in the biogas indicate that the landfill site is still active and the wastes contain readily biodegradable organic matter. Relatively low concentrations of BOD, COD and DOC may indicate not only the final stage of the landfill development. Their concentrations may also decrease as a result of natural biodegradation (natural attenuation mechanisms) and mixing (dilution). While isotopic data suggest a signature of the landfill leachate in the groundwater in the vicinity of the landfill site, chemical analyses do not show its strong influence, which could be indicative of natural attenuation mechanisms. It is evident that determination of $\delta^{13}C_{DIC}$ in the landfill leachate can provide a powerful tool for the assessment of the landfill activity and help to minimize the number of samples necessary for the analysis of conventional parameters.

Comprehensive data concerning the Otwock landfill site suggest that the intensity and direction of changes clearly correlate with water balance of the landfill body. This demonstrates the need for increasing the frequency of the monitoring above the one specified in the regulations on landfill sites. Annual measurements may be insufficient for an actual and reliable assessment of the environmental impact of the landfill site. The frequency and scope of the monitoring should be adapted to the local climate and hydrogeological conditions and the properties of the deposited waste.

Determination of $\delta^{13}C_{DIC}$ in the landfill leachate may provide a powerful tool for assessing the landfill activity in other sites and it can help in minimizing the number of samples collected for the analysis of conventional parameters.

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